Nonlinear magnetoelectric effect in a layered ferromagnetic-piezoelectric heterostructure excited by transverse magnetic field

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Nonlinear magnetoelectric effect in a heterostructure containing layers of amorphous ferromagnet FeBSiC and piezoelectric lead zirconate titanate ceramics has been investigated. The heterostructure was subjected to permanent, H_0, and alternating, h, magnetic fields applied in the structure plane. In contrast to previous studies, the excitation field was directed perpendicular to the permanent field ($h \perp H_0$). Generation of even voltage harmonics across the piezoelectric layer was observed for excitation fields in the range of 0-3 Oe. The dependence of the second harmonic amplitude on the permanent field strength was found to differ significantly from the similar dependence upon longitudinal excitation, $h // H_0$. A theory was developed, that describes the field dependence of voltage harmonic amplitudes for the magnetoelectric effect excitated by a transverse magnetic field.

Keywords: nonlinear magnetoelectric effect, heterostructure, magnetostriction, piezoelectricity, generation of harmonics.

Magnetoelectric (ME) effects in multiferroic single-phase materials and composite structures, performing mutual transformation of electric and magnetic fields, are intensively studied due to prospects of their use for designing highly sensitive magnetic field sensors, radio-signal processing devices, and new data storage magnetic elements [1-3]. In planar heterostructures consisting of ferromagnetic (FM) and piezoelectric (PE) layers, the ME effect results from a combination of magnetostriction of the FM layer and piezoelectricity in the PE layer due to mechanical coupling between the layers [4]. When the structure is exposed to a permanent magnetic field H_0 and an alternating magnetic field $h(f)$ of frequency f, the ME effect manifests itself as generation of an ac voltage $u(f)$ across the electrodes of the PE layer. It appears that with increasing the amplitude of the excitation field h, the nonlinearity of the FM layer magnetostriction dependence on the field λ(H) leads to a number of nonlinear effects, such as generation of voltage harmonics [5,6], frequency mixing [7,8], hysteresis suppression [9], and bistability [10].

In majority of works, the ME effect was studied in structures whose PE layer was poled perpendicular to the structure plane while both permanent and ac magnetic fields were applied in the plane of the structure in order to reduce the influence of demagnetization effects. Moreover, mostly "longitudinal" excitation of the ME effect by an ac field directed along a permanent field (i.e. h // H_0) was considered.

In several works [11–13], the linear ME effect was observed in layered FM-PE structures under "transverse" excitation, when both fields were applied in the structure plane, but the excitation field was directed perpendicular to the dc field, i.e. $h \perp H_0$. Recently generation of voltage harmonics was observed in the FM-PE structure excited by a rotating magnetic field [14]. The terms "longitudinal" and "transverse" excitation are commonly used to describe excitation of low-frequency oscillations of magnetization [15] and ferromagnetic resonance [16] in FM samples and therefore it is appropriate to use them for describing the ME effect too.

In this work, the nonlinear ME effect in the FM-PE layered structure subjected to a transverse excitation was observed and investigated in detail. It was found that the transverse excitation geometry results in a significant change of the nonlinear ME effect characteristics in comparison with the more traditional longitudinal excitation.

In the experiments, a bilayer structure schematically shown in Fig. 1 was used. The structure contained a FM layer of amorphous ferromagnet FeBSiC (Metglas 2605S3A) with the thickness of 20 μm and saturation magnetostriction of $\lambda_S \approx 21 \times 10^{-6}$ in saturation field $H_S \approx 100$ Oe and a PE layer of lead zirconate titanate ceramics $PbZr_{0.52}Ti_{0.48}O_3$ (PZT) with the thickness of a_p = 300 μm and piezoelectric coefficient of $d_{31} \approx 175$ pm/V. An amorphous ferromagnet was chosen because it has the largest nonlinear piezomagnetic coefficient and can be saturated by a low magnetic field. The Ag-electrodes, ~2 μm thick, were deposited on the PZT layer that was poled perpendicular to its plane in permanent electric field E =10 kV/cm. The layers were bonded using epoxy adhesive.

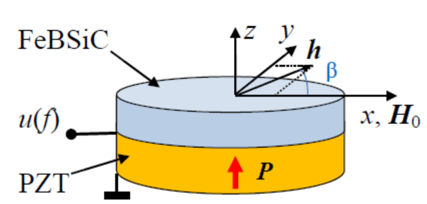

Fig.1 Schematic view of the studied ferromagnetic-piezoelectric heterostructure. The arrows indicate the direction of piezoelectric layer polarization P together with directions of permanent H_0 and alternating h magnetic fields for excitation of the ME effect by transverse field.

To reduce the effect of demagnetization, the structure had the shape of a disk with the diameter of 15 mm. A permanent magnetic field H_0 = 0 -100 Oe and ac excitation magnetic field $h\cos(2\pi ft)$ with the amplitude up to h = 0-3 Oe and frequency of f = 0-25 kHz were applied using two Helmholtz coils. Both fields, H_0 and h, were oriented in the structure plane. In general case, the ac field h was dierected at the angle β with respect to the H_0 field, as shown in Fig. 1. To study the transverse excitation of the ME effect, the ac field was directed along the y axis, perpendicular to the permanent field, $h \perp H_0$. For the sake of comparison, the longitudinal excitation was investigated too. For that the ac field was applied along the x axis, parallel to the permanent field, $h // H_0$. The spectrum of the voltage response generated by the structure, $u(f)$, was recorded using a SR770 analyzer for various H_0 and h values.

Figure 2 shows a typical frequency spectrum of voltage generated by the FeBSiC-PZT structure when excited with a transverse ($h \perp H_0$) ac field of amplitude h = 2 Oe and frequency f_0 = 5 kHz at H_0 ≈11 Oe. The frequency f_0 is chosen so that it and its harmonics do not coincide with the frequencies of acoustic resonances of the structure. In addition to the main voltage harmonic u_1 of frequency 5 kHz, the second u_2, third u_3, and fourth u_4 voltage harmonics of frequencies $f_n = f_0 n$, where n = 1.2 ..., appear in the spectrum. This indicates the high nonlinearity of the ME effect. The specific feature of the spectrum (as compared with the longitudinal excitation) is a large amplitude of even harmonics. In the frequency range of 2-3.5 kHz and near the frequency ~7.5 kHz one can see small peaks corresponding to generation of voltage at frequencies of lower flexural vibration modes of the structure.

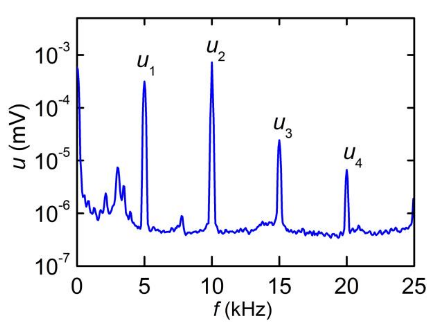

Fig. 2 Frequency spectrum of the voltage generated by the FeBSiC-PZT structure under transverse excitation of the ME effect by ac magnetic field with frequency f_0=5 kHz and amplitude h=2 Oe.

Figure 3 shows second harmonic amplitude u_2 as a function of permanent field H_0 for transverse (curve 1) and longitudinal (curve 2) excitations by an ac field of frequency 5 kHz and amplitude h = 2 Oe. As can be seen, these dependences differ significantly. For transverse excitation, $u_2 \approx$ 0.45 mV at H = 0 and the harmonic amplitude initially increases with H_0, then reaches its maximum at H_1 ≈11 Oe and after that it gradually decreases as the FM layer becomes saturated. In the case of longitudinal excitation, u_2 also initially increases, reaching its maximum at the same field $H_1 \approx 11$ Oe, but then it drops to zero at characteristic field $H_m \approx 21$ Oe. After that it passes through a local maximum, and then again tends to zero as the FM layer is saturated. Note, that the voltage u_2 changes the phase on π when the field passes through H_m. The maximum amplitudes of the second harmonics are approximately the same for both types of excitation. The efficiency of the second harmonic generation under transverse magnetic field excitation reached $\alpha_E^{(2)} = u_2/(a_p h^2) \approx$ 6.2 mV/(cm x Oe^2). The inset in Fig.3 shows the dependence of the second harmonic amplitude u_2 on the excitation field amplitude h for the transverse excitation at $H_1 \approx 11$ Oe. It is seen, that in the small amplitude region h <3 Oe, the dependence is well approximated by a parabola $u_2 = bh^2$ with $b = 0.185$ mV/Oe^2.

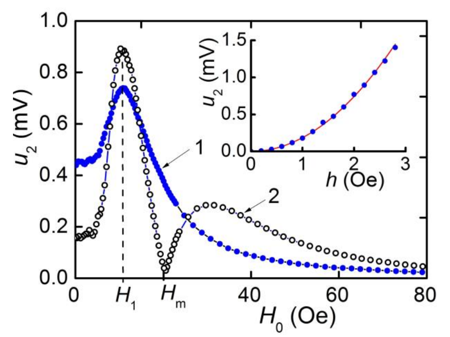

Fig. 3 Second harmonic amplitude as a function of permanent magnetic field for the ME effect excited by transverse (curve 1) and longitudinal (curve 2) ac magnetic fields. The inset shows the dependence of u_2 on the amplitude of ac field for transverse excitation at H_0=11 Oe. Solid line here corresponds to a quadratic approximation.

The obtained results can be explained using a model, in which the nonlinear ME effects in FM-PE structures occur entirely due to a nonlinear dependence of magnetostriction λ on magnetic field H [17]. Let us assume that the FM and PE layers of the structure are uniform and isotropic. It can be shown that when a magnetic field H is applied, the heterostructure would generate voltage $u(H) = Ad_{31}\lambda(H)$, where A is a constant that depends only on dimensions and dielectric parameters of the layers, d_{31} is the longitudinal piezoelectric module of the PE layer and $\lambda(H)$ is the magnetostriction of the FM layer at this field. Taking the field acting on the structure in the form $H = H_0 + \delta H \quad (\delta H << H_0)$ and expanding $\lambda(H)$ as a Taylor series in the vicinity of H_0, the following relation can be obtained:

$$u = Ad_{31}[\lambda(H_0) + \lambda^{(1)}\delta H + \frac{1}{2}\lambda^{(2)}(\delta H)^2 + ...], \tag{1}$$

where $\lambda^{(1)} = \partial\lambda/\partial H\big|_{H_0}$, $\lambda^{(2)} = \partial^2\lambda/\partial H^2\big|_{H_0}$ are the first (piezomagnetic coefficient) and the second (nonlinear piezomagnetic coefficient) derivatives of magnetostriction with respect to the field at $H=H_0$, respectively. Obviously, that for an isotropic structure, the coefficients $\lambda^{(1)}$ and $\lambda^{(2)}$ do not depend on the direction of the H-field in the plane of the structure. At the same time, the magnitude of the field variation δH depends on the mutual orientation of the h and H_0 fields. For an arbitrary orientation of h with respect to H_0 (see Fig. 1) and small amplitude of the excitation field $h << H_0$, we have $\delta H = \sqrt{H_0^2 + h^2 + 2H_0 h\cos\beta} - H_0 \approx h^2/(2H_0) + h\cos\beta$.

When the ME effect is excited by a transverse field ($\beta \approx \pi/2$), $h \perp H_0$, the total field changes by $\delta H \approx H^2/(2H_0)$. For the longitudinal excitation ($\beta \approx 0$), h // H_0, the total field change is $\delta H \approx h$. Substitution of δH and $h\cos(2\pi ft)$ into (1) gives the following relation for the voltage generated by transverse excitation of the ME effect

$$\begin{aligned} u \approx Ad_{31}\left[\lambda(H_0) + \frac{1}{4}\frac{\lambda^{(1)}}{H_0}h^2\right] + \frac{1}{4}Ad_{31}\frac{\lambda^{(1)}}{H_0}h^2\cos(4\pi ft) \\ + \frac{1}{64}Ad_{31}\frac{\lambda^{(2)}}{H_0^2}h^4\cos(8\pi ft) + \ldots \end{aligned} \tag{2}$$

It follows from (2), that in the case of transverse excitation, the structure should generate a permanent voltage (the first term) and even voltage harmonics. The permanent voltage rapidly decreases due to the finite conductivity of the PE layer, therefore it will not be taken into account in the further consideration. The dependence of the second harmonic amplitude on field H_0 and excitation field h is determined, as it is seen from (2), by field dependence of the product

$$u_2(H_0, h) \sim [\lambda^{(1)}(H_0)/H_0]h^2. \tag{3}$$

In a similar way, one can obtain that at longitudinal excitation of the ME effect, the structure will generate both odd and even voltage harmonics. In this case, the dependence of the first and the second harmonic amplitudes on permanent field H_0 and excitation field h is determined, respectively, by functions [16]

$$u_1(H_0, h) \sim \lambda^{(1)}(H_0)h \quad \text{and} \quad u_2(H_0, h) \sim \lambda^{(2)}(H_0)h^2. \tag{4}$$

Note that for transverse excitation of the ME effect (see Fig. 2) the voltage spectrum, in addition to even harmonics, also contains odd harmonics u_1 and u_3. However, our measurements showed, that a significant contribution to the voltage u_1 arises due to a direct electromagnetic pickup which does not depend on H_0. In addition, generation of the odd harmonics u_1 and u_3 can be also caused by anisotropy of the FM layer, which was not controlled in our measurements. The magnetic anisotropy field H_a can violate the condition $h \perp H_0$ and lead to a noticeable contribution of the longitudinal effect to the odd harmonic generation. Also, the ME effect due to shear deformations can lead to a generation of the main harmonic [18].

To confirm predictions of the theory regarding the specific dependence of the harmonics amplitudes at transverse and longitudinal excitation on the permanent magnetic field, additional measurements of magnetostriction of the FeBSiC layer had been carried out. The measurements were performed on a stand-alone disk sample, 15 mm in diameter, by the tensometric method with an accuracy of $\Delta H \approx 0.1$ Oe and $\Delta\lambda \approx 0.5\text{x}10^{-6}$ using an automated setup [19].

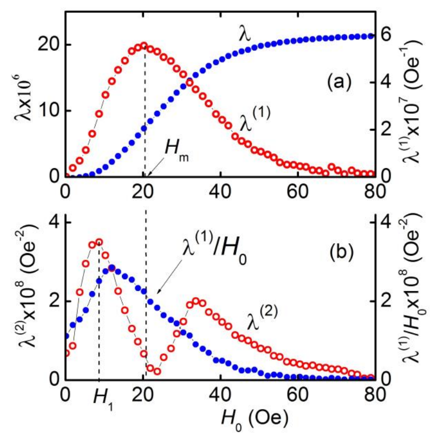

Fig. 4 (a) Measured dependence of magnetostriction λ and calculated dependence of piezomagnetic coefficient $\lambda^{(1)}$ on permanent magnetic field H_0 for FeBSiC layer. (b) Calculated nonlinear piezomagnetic coefficient $\lambda^{(2)}$ and ratio $\lambda^{(1)}/H_0$ as a function of dc field H_0. Solid lines are drawn to guide the eye.

The measured $\lambda(H_0)$ dependence is shown in Fig. 4a. The same figure shows field dependence of piezomagnetic coefficient $\lambda^{(1)}(H_0)$, calculated by numerical differentiation of the measured curve $\lambda(H_0)$. The $\lambda^{(1)}(H_0)$ dependence shows a maximum at field $H_m \approx 21$ Oe. This leads to a maximum in the field dependence of amplitude of the first ME voltage harmonic in the case of longitudinal excitation [20].

Figure 4b displays field dependence for the ratio $\lambda^{(1)}(H_0)/H_0$, calculated by the same method. This function, according to (3), determines the shape of the field dependence for the second harmonic amplitude under transverse excitation of the ME effect. Comparison of the $\lambda^{(1)}(H_0)/H_0$ curve in Fig. 4 and curve 1 in Fig. 3 proves that the theory well describes the experimental observatios. Figure 4b also shows the field dependence of the nonlinear piezomagnetic coefficient $\lambda^{(2)}(H_0)$, obtained by double differentiating the experimental $\lambda(H_0)$ curve. Comparison of this curve with curve 2 in Fig. 3 confirms that the theory describes well the dependence of the second harmonic amplitude on the field strength for longitudinal excitation of the ME effect [17]. The proposed theory, as can be seen from the inset in Fig. 3, also describes the quadratic dependence of the second harmonic amplitude u_2 on the ac field amplitude h upon transverse excitation of the ME effect.

The results of measurements (Fig. 3) and calculations (Fig. 4b) enabled us to conclude, that the shape of the dependence $u_2(H_0)$ can be controlled by changing the angle between the excitation field h and permanent field H_0 from zero to 90^0. It should be also noted, that the decrease in the second harmonic amplitude in the low field region $H_0 \approx 0$ indicates deviation of the real field dependence of the FeBSiC layer magnetostriction from the quadratic law, which was observed for other magnetostrictive materials [21].

Thus, the nonlinear ME effect in a tangentially magnetized bilayer ferromagnetic-piezoelectric heterostructure was observed and investigated upon excitation by ac magnetic field directed perpendicularly to the permanent field. It is shown, that in the case of transverse ME effect excitation, mostly even voltage harmonics are generated and the dependence of the second harmonic amplitude on the permanent field differs significantly from the similar dependence upon longitudinal excitation. The amplitude of the second harmonic gradually decreases with increasing the permanent field and quadratically depends on the excitation field amplitude. A theory is developed that describes well the field dependences of the ME effect characteristics when it is excited by a transverse field.

The results obtained demonstrate strong anisotropy of the nonlinear ME effect excitation and should be taken into account when designing magnetic field sensors and radio-signal processing devices based on the nonlinear ME effect in layered ferromagnetic-piezoelectric structures.

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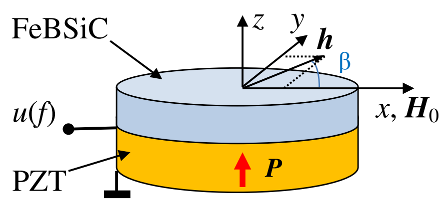
FeBSiC
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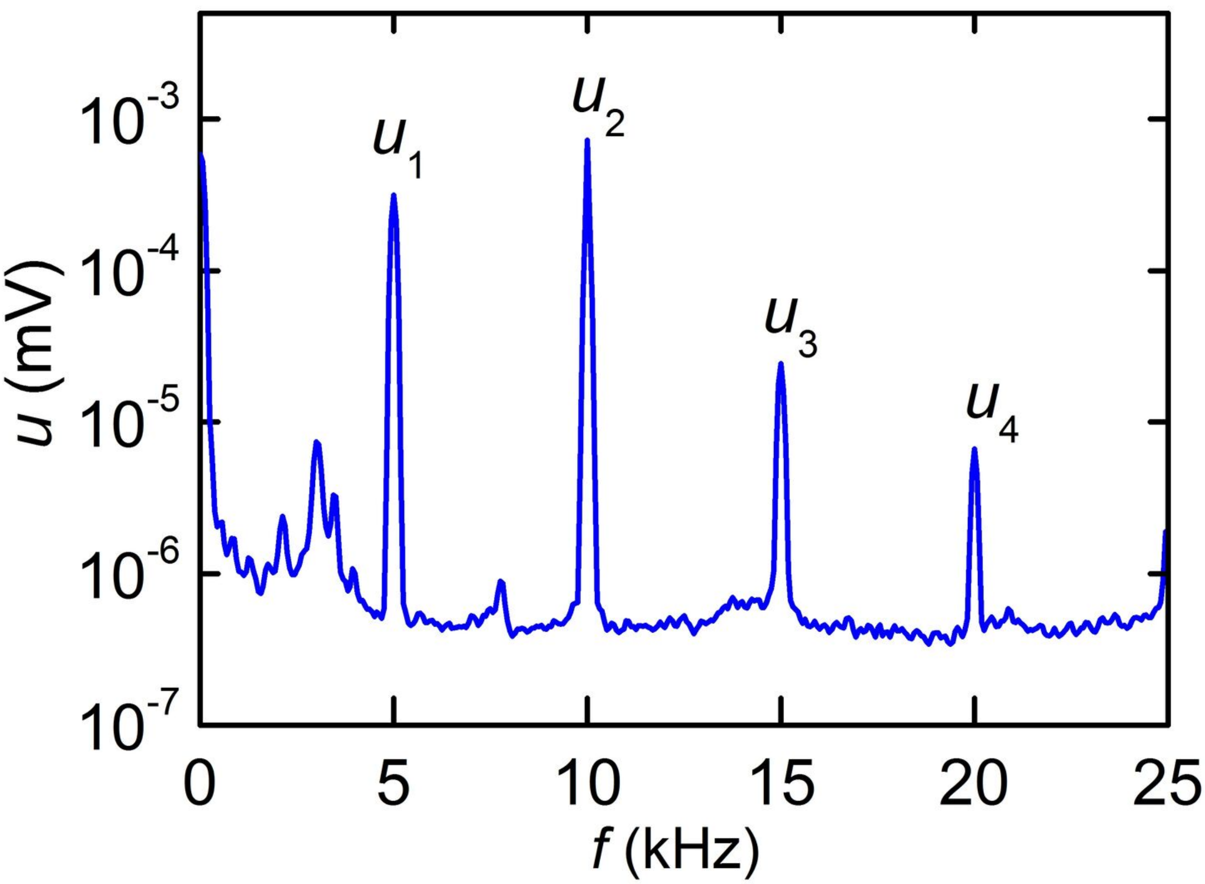
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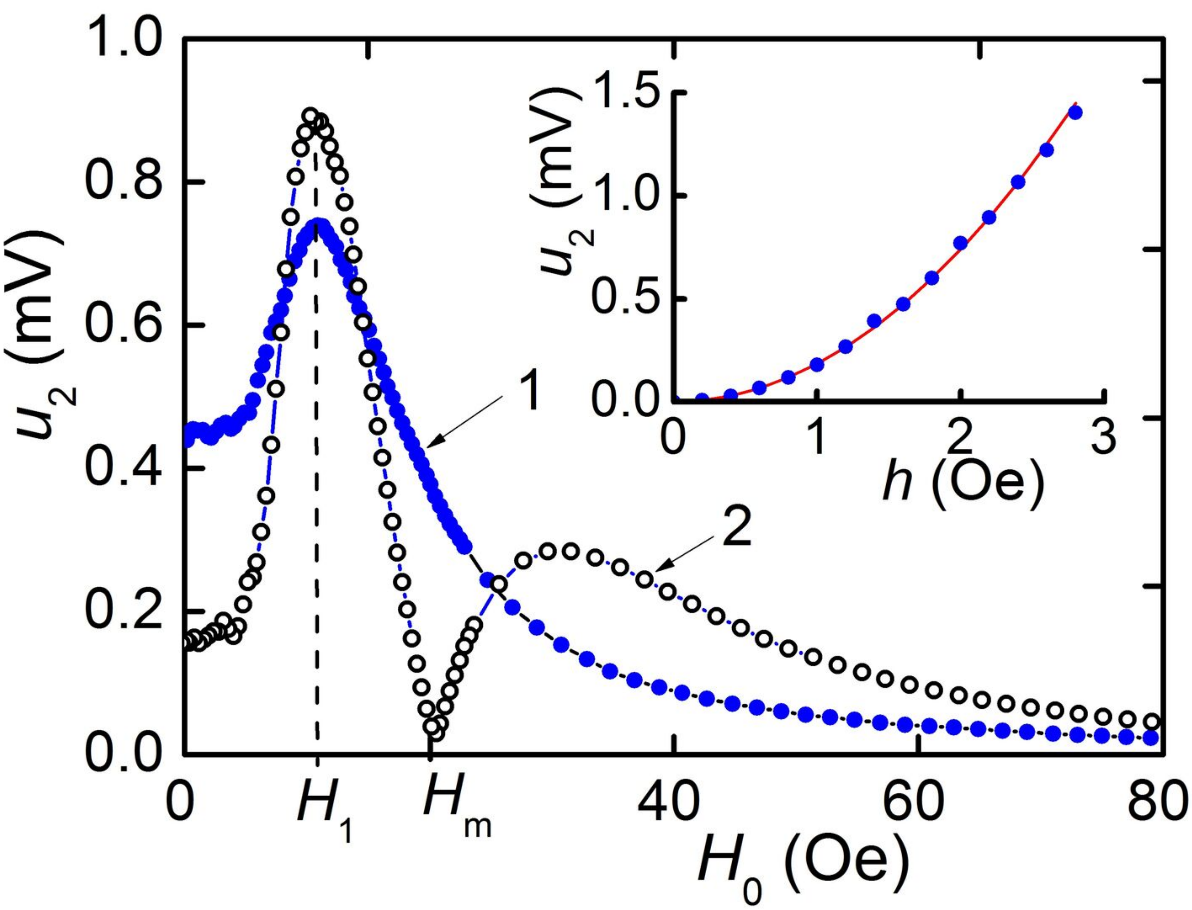
1.0
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